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CURABLE INSULATION MATERIALS, ELECTRICAL WIRES COATED THEREWITH,  
AND THE PRODUCTION METHOD OF ELECTRICAL WIRES

Author (Katsutoshi Takahashi)

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Inventor : Yukata Hashimoto, June Shiraga,  
Masayuki Kamei

Applicant : Dainippon Ink Kagaku Kogyo Co

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Native Title : 「硬化性絶縁性材料、それで被覆された  
電 線及び電線の製造方法」

English Title : Curable insulation materials,  
electrical wires coated  
therewith, and the production  
method of electrical wires

## Specification

### 1. [Title of the invention]

CURABLE INSULATION MATERIALS, ELECTRICAL WIRES COATED THEREWITH  
AND THE PRODUCTION METHOD OF ELECTRICAL WIRES

### 2. [Scope of the patent claims]

(1). Curable insulation material which contains polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer or polyfluoroalkyl group - containing monoepoxy monomer.

(2). Curable insulation material described in patent claim item 1 which, furthermore, contains polyfluoroalkyl group or polyfluoroalkylene group - containing polymer.

(3). Curable insulation material which consists of polyfluoroalkyl group - containing mono alpha, beta- unsaturated ethylene monomer and poly alpha, beta -unsaturated ethylene monomer.

(4). Curable insulation material described in patent claim item 3 wherein poly alpha, beta - unsaturated ethylene monomer is polyfluoroalkyl group or polyfluoroalkylene group - containing poly alpha, beta- unsaturated ethylene monomer.

(5). Curable insulation material described in patent claim item 3 or 4 which, furthermore, contains the polymer of polyfluoroalkyl group or polyfluoroalkylene containing mono alpha, beta - unsaturated ethylene monomer.

(6). Curable insulation material described in patent claim item 5 wherein the polymer of polyfluoroalkyl group or

polyfluoroalkylene group - containing mono alpha, beta - unsaturated ethylene monomer is the polymer of the corresponding monomer.

(7). Curable insulation material which consist of polyfluoroalkyl containing monoepoxy monomer and polyepoxy monomer.

(8). Curable insulation material described in patent claim item 7 wherein polyepoxy monomer is either polyfluoroalkyl group or polyfluoroalkylene containing polyepoxy monomer.

(9). Curable insulation material described in patent claim item 7 or 8 that, furthermore, contains the polymer of polyfluoroalkyl group or polyfluoroalkylene group - containing monoepoxy monomer.

(10). Curable insulation material described in patent claim item 9 wherein the polymer of polyfluoroalkyl group or polyfluoroalkylene group - containing monoepoxy monomer is the polymer of corresponding monomer.

(11). It is the insulation coated electrical wire coated by the cured product of the curable insulation material in claim item 1

(12). The production method of insulation coated electrical wire characterized such that the curable insulation material described in claim item 1 is coated on electrical wire conductive body, then, by irradiating the active energy beams, said material is cured.

### 3. [Detailed explanation of the invention]

#### [Utilized field in industry]

The present invention relates to curable insulation material, electrical wires coated therewith, and the production method of electrical wires.

#### [Prior arts]

In all of Japan Patent Disclosure Hei1-249744 gazette, European disclosure 309783 gazette, and European disclosure 322624 gazette, are described the curable insulation material consisting of poly(meta) acrylate monomer which contains fluorine atoms.

#### [Problems the present invention attempts to solve]

However, regarding the electrical wires coated by the cured product of the curable insulation material described in above described publications, its insulation performance was insufficient.

#### [Means to solve the problems]

The inventors herein studied very hard in view of the above described situation, then, found out that the electrical wires coated by the curable insulation material were superior in insulation performance wherein polyfluoroalkyl group - containing mono(meta) acrylate monomer or polyfluoroalkyl group - containing monoepoxide monomer was added into the traditional curable insulation material described above, thus completing the present invention.

That is, this provides the production method of curable insulation material characterized such that curable insulation material contains polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer or polyfluoroalkyl group - containing monoepoxy monomer,

As to the insulation coated electrical wires coated by the cured product of said curable insulation materials, the curable insulation material is coated on the on the electrical wire conductive body, then, by irradiating the active energy beams, said material is cured.

In the present invention, polyfluoroalkyl group - containing mono alpha, and beta - unsaturated ethylene monomer means that it contains one polymerized alpha, beta unsaturated ethylene group, and it is the monomer which contains at least one polyfluoroalkyl group, and polyfluoroalkyl group - containing monoepoxy monomer means that it has one polymerizable epoxy group and it is a monomer that has at least one polyfluoroalkyl group.

This polymerizable monomer refers to what combines one with heat curable characteristics and one with active energy beams curable characteristics.

As to the content amount of the fluorine atoms contained in the entire weight of insulation material of the present invention, from its insulation performance stand point, it is usually 3 weight% or more, preferably 10 weight% or more.

Regarding polyfluoroalkyl group - containing mono'alpha, beta - unsaturated ethylene monomer which is contained as essential ingredient in the insulation material of the present invention, its curing progresses by a radical polymerization mechanism, and regarding polyfluoroalkyl group - containing monoepoxy monomer, its curing progresses by cation polymerization mechanism and it can be a joint system of radical polymerization mechanism and cation polymerization mechanism.

Next, radical polymerization system and cation polymerization system will be explained in sequence.

In case of radical polymerization system, the insulation material of the present invention is mainly comprised of above described polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer, and depending on needs, mono alpha, beta - unsaturated ethylene monomer or poly alpha, beta - unsaturated ethylene monomer without polyfluoroalkyl group is used.

In case of radical polymerization system, the polymerized group of polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer, mono alpha, beta -unsaturated ethylene monomer and poly alpha, beta- unsaturated ethylene monomer that do not contain polyfluoroalkyl group are not particularly limited as long as they are alpha, beta- unsaturated ethylene group,

However, from the stand point of polymerization curing characteristics, polymerizable monomer which contains methacryloil group, acryloil group, halo acryloil group or cyano acryloil group or alpha, beta- unsaturated ethylene containing ester dicarboxylate is preferred.

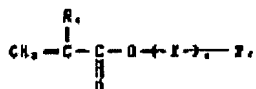
In the following, unless otherwise stated, methacryloil group, acryloil group, halo acryloil group or cyano acryloil group are generically called [acryloil group], the polymerizable monomer which contains methacryloil group, acryloil group, halo acryloil group or cyano acryloil group are generically called [acryloil group - containing polymerizable monomers] generically, and methacrylate, acrylate, halo acrylate and cyano acrylate are called [acrylate] generically.

The [(Meta) acrylate) in the compound shown in examples are assumed to represent all of methacrylate, acrylate, halo acrylate or cyano acrylate.

As polyfluoroalkyl group - containing mono alpha, beta-unsaturated monomer, polymerizable monomer that contains, for instance, polyfluoroalkyl group in molecules and also one acryloil group (hereafter called fluorine containing monoacrylate ) (1-a) can be listed.

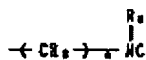
Fluorine containing monoacrylate (1-a) is as follows: for instance, perfluoro alkyl group or partially fluoro aliphatic group with carbon number of 1~20 is bonded with acryloil group

The following compound expressed by the following general formula:

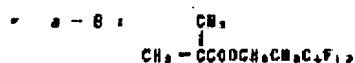
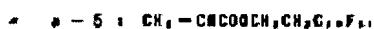
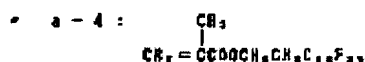
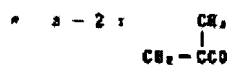

$$\text{---}(\text{CF}_2\text{CFO})\text{---CF}(\text{CF}_3)\text{---}$$

|  
CF<sub>3</sub>

等でも良く、R<sub>1</sub>はH、

$$-\text{CH}_2\underset{\text{OH}}{\text{CH}}-(\text{CH}_2)-$$

$$\begin{array}{c} \text{CH}_3 - \text{C} - \text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5 \dots \\ | \\ \text{CH}_2\text{CH}_2\text{C}_6\text{F}_5 \dots \end{array}$$

Or as shown in the following formula,

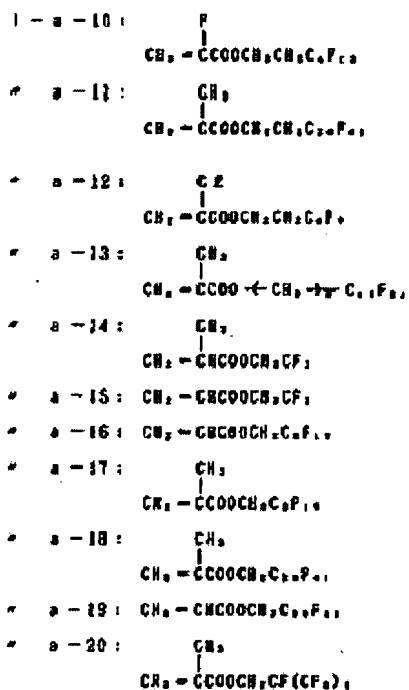


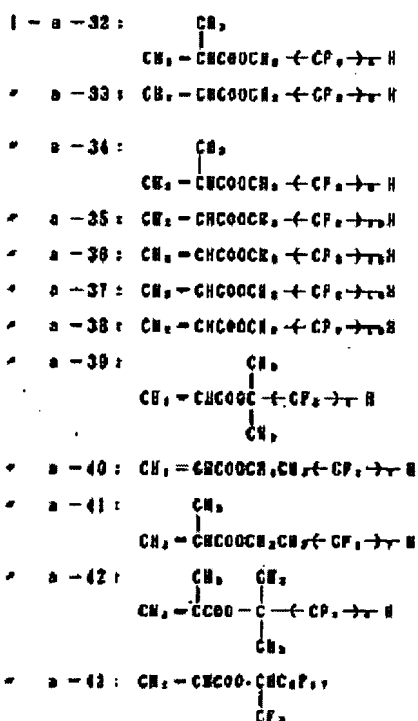
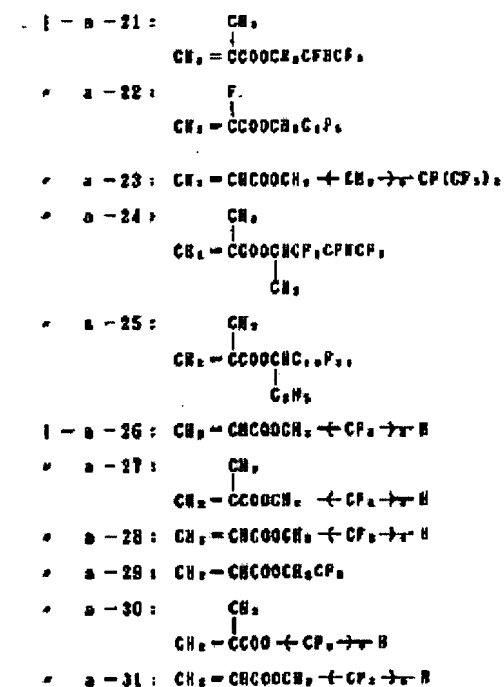
8

perfluoroalkyl groups in the molecules

(In the formula, "l or el" is an integer of 1-14).

As these specific examples, there are followings.





Fluorine containing monoacrylate (1-a) can be the mixture of the compounds of two types or more of different structures. And,

of course, the present invention is not limited by above described specific examples, and as described later, polyfluoro alkyl containing alpha, beta - unsaturated ethylene ester dicarboxylate can be used.

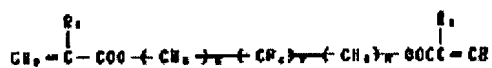
As the polymerizable monomer which can be combined with above described polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer, there is, for instance, poly alpha, beta - unsaturated ethylene monomer.

In the present invention, poly alpha, beta- unsaturated ethylene monomer refers to the monomer which contains at least two polymerizable alpha, beta- unsaturated ethylene groups.

Poly alpha, beta - unsaturated ethylene monomer can be largely classified into polyfluoroalkyl group or polyfluoroalkylene group - containing poly alpha, beta unsaturated ethylene monomer, and Poly alpha, beta - unsaturated ethylene monomer without polyfluoroalkyl group or polyfluoroalkylene group.

Among those, as polyfluoroalkyl group or polyfluoroalkylene group - containing poly alpha, beta - unsaturated ethylene monomer, there are for instance polymerizable monomer which contains for instance, polyfluoroalkyl group or polyfluoroalkylene group and also, contains at least two acryloil group (hereafter, called fluorine containing multifunctional acrylate) (1-b).

As the fluorine containing multifunctional acrylate (1-b), compounds can be listed with following general formula,

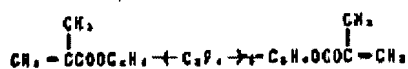


(In the formula, X is an integer of

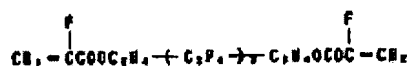
1~2, Y is an integer of 4~12, R1 is same as before). As the specific compound for them, the following can be exemplified.



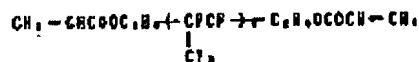
(a, b are 1~4)



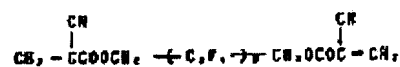
b = 4



b = 5



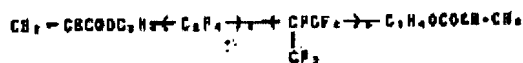
b = 6



b = 7



b = 8



(a, b are 1~4)

[In the formula, R1 is same as before,

However, X and Y are same as before) (1-b-9)

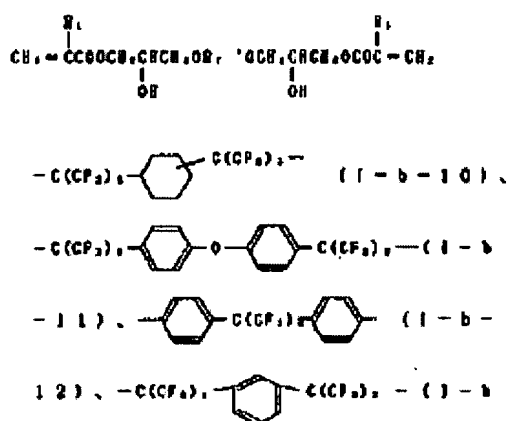
As other fluorine containing multifunctional acrylate, the compounds represented by the following formula can be listed, that are described in Japan Patent Disclosure Hei 1-249744 gazette, European Disclosure 309783 gazette, same 322624 gazette.

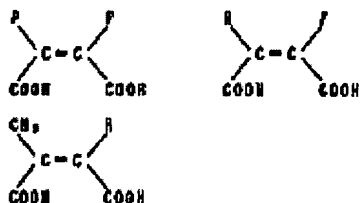
Regarding fluorine containing multifunctional acrylate (1-b)

along with fluorine containing monoacrylate, it is important that they manifest insulation performance and provide flame retardation property and also manifest the mechanical strength to the extent of being practical. And, of course, the present invention is not limited to above described specific special examples.

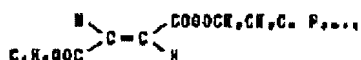
Alpha, beta - unsaturated ethylene dicarboxylic acid or its ester can be largely classified into polyfluoroalkyl group - containing one and one without polyfluoroalkyl, and polyfluoroalkyl group - containing Alpha, beta - unsaturated ethylene dicarboxylic acid or its ester is polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer which is the essential ingredient of the insulation material of the present invention.

As the alpha, beta - unsaturated ethylene dicarboxylic acid, one without polyfluoroalkyl group, for instance, the following can be listed.





As the  $\alpha,\beta$  - unsaturated ethylene ester dicarboxylate, for instance, general formula (II) is a compound expressed as follow:



[in the formula, R<sub>3</sub> and R<sub>4</sub> are H, F, Cl, CN or CH<sub>3</sub>, and R<sub>3</sub> and R<sub>4</sub> can be same or different, and R<sub>5</sub> and R<sub>6</sub> are the group that contains H or alkyl group or fluoro alkyl group carbon number 1~20, and R<sub>5</sub> and R<sub>6</sub> can be same or different, however, if one of them is H, other is a group other than H].

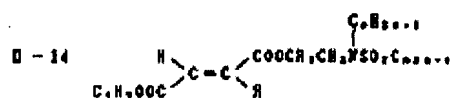
As the specific compounds, there are following ones combining one with polyfluoroalkyl and one without that.

- 0-1: diethyl ester fumarate
- 0-2: di-n-butyl ester fumarate
- "-3: di-iso-butyl ester fumarate
- "-4: di-t-butyl ester fumarate
- "-5: dipropyl ester fumarate
- "-6: per-2-ethyl hexyl ester fumarate
- "-7: dihexyl ester fumarate
- "-8: ethyl butyl ester fumarate
- "-9: dibutyl ester maleate
- "-10: dioleyl ester maleate

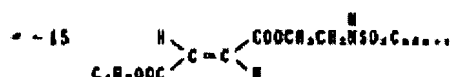
"-11: n-butyl fumarate, t-butyl ester

"-12: ethyl fumarate, iso-butyl ester

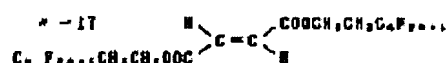
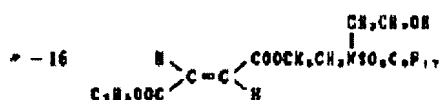
"-13:



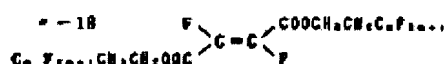
(p は 1 ~ 20 の整数、m は前記と同様)



(m は前記と同様)

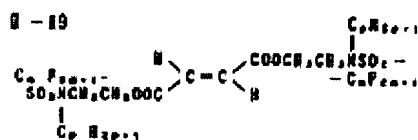


(m は前記と同様)

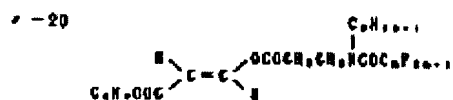


(m は前記と同様)

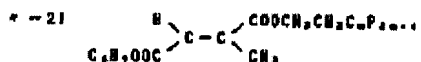
(m is an integer 1~18)



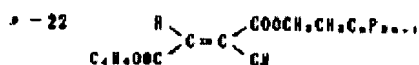
(p および m は前記と同様)



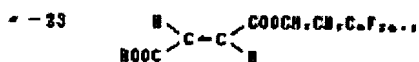
(p および m は前記と同様)



(m は前記と同様)



(m は前記と同様)



(m は前記と同様)

First chemical structure

(p is an integer 1~20, m is same as before)

**Second chemical structure**

(m is same as before)

**Third chemical structure**

(m is same as before)

**Fourth chemical structure**

(m is same as before)

**First one**

(p and m are same as before)

**Second one**

(p and m are same as before)

**Third one**

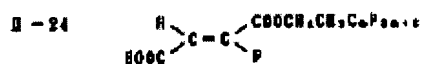
(m is same as before)

**Fourth one**

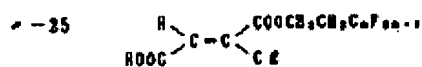
(m is same as before)

**Fifth one**

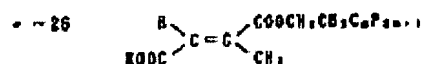
(m is same as before)



(m は前記と同義)



(m は前記と同義)

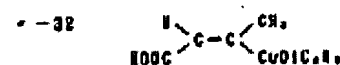
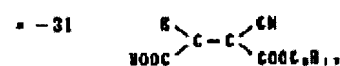
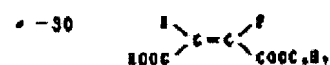


(m は前記と同義)

m-27 フマル酸モノ-n-ブチルエステル

m-28 フマル酸モノ-n-エチルヘキシル  
エステル

m-29 マレイン酸モノオレイルエステル



First one

(m is same as before)

Second one

(m is same as before)

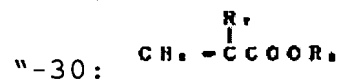
Third one

(m is same as before)

"-27: mono-n-butyl ester fumarate

"-28: mono-n-ethyl hexyl ester fumarate

"-29: monooleyl ester maleate



"-31: same

"-32: same

Alpha, beta - unsaturated ethylene dicarboxylic acid or its ester (II) is very important in manifesting mechanical strength and flexibility of the insulation material obtained by curing the insulation material of the present invention.

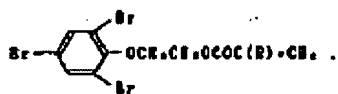
As the polymerizable material of the present invention, other than above described each monomer group, monoacrylate without polyfluoroalkyl group and multi functional acrylate without polyfluoroalkyl group can be used favorably.

As the monoacrylate without polyfluoroalkyl group, the compound represented by the following general formula can be listed

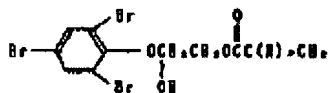
(In the formula, R is H, F, Cl, CN or CH<sub>2</sub>, and R' is a group with carbon number 1-20), as their specific examples, the following compounds can be listed;

Methyl (meta) acrylate, ethyl (meta) acrylate, n-propyl (meta) acrylate, i-propyl (meta) acrylate, n-butyl (meta) acrylate, 1-butyl (meta) acrylate, t-butyl (meta) acrylate, hexyl (meta) acrylate, octyl (meta)acrylate, decil (meta) acrylate, isodecil (meta) acrylate, lauryl (meta) acrylate, stearyl (meta) acrylate, and isostearyl (meta) acrylate and the like that are aliphatic group ester (meta) acrylate; glycerol (meta) acrylate, 2-hydroxy propyl (meta) acrylate, 3-chloro-2- hydroxy propyl (meta) acrylate, glycidyl (meta) acrylate, allyl (meta) acrylate, butoxy ethyl (meta) acrylate, butoxy ethylene glycol (meta) acrylate, N, N-diethyl amino ethyl (meta) acrylate, N-N- dimethyl amino ethyl

(meta) acrylate, 2-ethyl hexyl (meta) acrylate,  
 gamma?-methacryloxy propyl trimethoxy silane, 2-methoxy ethyl  
 (meta) acrylate, methoxy diethylene glycol (meta) acrylate,  
 methoxy dipropylene glycol (meta) acrylate, nonyl phenoxy  
 polyethylene glycol (meta) acrylate, nonyl phenoxy polypropylene  
 glycol (meta) acrylate, alonix M-5700 (made by Toa gousei kagaku  
 Co), phenoxy ethyl (meta) acrylate, phenoxy propylene glycol  
 (meta) acrylate, phenoxy dipropylene glycol (meta) acrylate,  
 phenoxy polypropylene glycol (meta) acrylate, and AR-260, MR-260,  
 AR-200, AR-204, AR-208, MR-200, MR-204, MR-208 (those made by  
 Daihachi Kagaku Co) that are ester phosphate group - containing  
 (meta) acrylate, furthermore, biscoat 2000, biscoat 2308 (these  
 made by Ohsaka YuKi Co), polybutadiene (meta) acrylate,  
 polyethylene glycol (meta) acrylate, polypropylene glycol (meta)  
 acrylate, polyethylene glycol- polypropylene glycol (meta)  
 acrylate, polyethylene glycol- polybutylene glycol (meta)  
 acrylate, polystyryl ethyl (meta) acrylate, and rite ester HOA-MS,  
 rite ester HOMS (made by Kyoei YUshi Co), benzyl (meta)acrylate,  
 cyclohexyl (meta) acrylate, dicyclopentenyl (meta) acrylate,  
 Dicyclopentenyl (meta) acrylate, isobornyl (meta) acrylate,  
 methoxy cyclodeca triene (meta) acrylate, phenyl (meta) acrylate;  
 and FA-512A, FA-512M made by Hitachi Kagaku Co; gamma-methacryloxy  
 propyl methyl dimethoxy silane, gamma acryloxy propyl trimethoxy  
 silane, gamma- acryloxy propyl methyl dimethoxy silane,



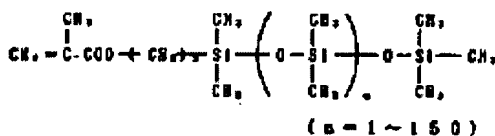
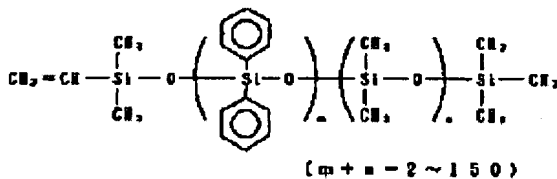
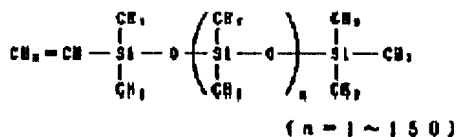
(R is H, CH<sub>2</sub>, Cl F or CN)



(R is H, CH<sub>2</sub>, Cl F or CN)

The monoacrylate without polyfluoroalkyl group is called diluted monomer by the industry which handles curable resin usually. And the insulation material of the present invention is used as the viscosity adjustment and is important in adjusting the mechanical strength and bending characteristics of cured product of the insulation material obtained by curing.

As the diluted monomer of the present invention, other than above described none - fluorine containing monoacrylate, there are styrene, P-methoxy styrene, dimethyl vinyl methoxy silane, dimethyl vinyl ethoxy silane, vinyl trichloro silane, vinyl trimethoxy silane, vinyl triethoxy silane, methyl vinyl dimethoxy silane, acrylic acid, methacrylic acid, and



And the like that are silicone chain containing vinyl compounds. And, of course, the present invention is not limited to the above described special examples.

Multifunctional acrylate (IV) without polyfluoroalkyl group of the present invention includes ones called multifunctional acrylate or special acrylate in the industry and one called pre-polymer, base resin, and oligomer or acryl oligomer, specifically the following can be exemplified.

- (i) polyhydric (meta) acrylate in which two units or more of (meta) acrylic acid are bonded with polyhydric alcohol,
- (ii) Polyester polyole obtained by reacting polyhydric alcohol and polybasic acid is polyester acrylate in which two units or more of (Meta) acrylic acid are bonded.

As the polyhydric alcohol in above described (i) and (ii), there

are ethylene glycol, 1,4-butane diol, 1,6- hexane diol, diethylene glycol, trimethylol propane, dipropylene glycol, polyethylene glycol, polypropylene glycol, penta erythrytol, dipenta erythrytol and the like. And for polybasic acid, there is phthalic acid, adipic acid, maleic acid, trimellitic acid, Itaconic acid, succinic acid, terephthalic acid, alkenyl succinic acid and the like.

(iii). as the functional group in which epoxy group of epoxy resin is esterified by (meta) acrylic acid, epoxy denatured (meta) acrylate is made into (meta) acryloil group.

As epoxy resin, there is bisphenyl A -epichloro hydrine type, phenol novolak - epichloro hydrine, polyhydric alcohol epichlorohydrine type lipid ring resins and the like.

(iv). Polyurethane acrylate which is obtained by reacting the hydroxyl group - containing (Meta) acrylate with polyhydric isocyanate compound.

As the polyhydric isocyanate compound, one is structured such that the center part of the molecules has polyester, polyether, polyurethane and the like, and on both ends isocyanate group is contained.

(v). In addition, there are polyether (meta) acrylate, melanine (meta) acrylate, alkyd (meta) acrylate, isocyanurate (meta) acrylate, silicon (meta) acrylate and the like.

As specific compound examples, there are followings:

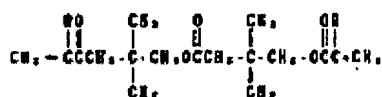
IV-1: ethylene glycol di(Meta) acrylate  
 IV-2: diethylene glycol di(meta) acrylate  
 IV-3: triethylene glycol di(meta) acrylate  
 IV-4: polyethylene glycol di(mea) acrylate (number average  
 molecular weight 150~1,000)

IV-5: propylene glycol di(meta) acrylate  
 IV-6: dipropylene glycol di(meta) acrylate  
 IV-7: tripropylene glycol di(meta) acrylate  
 IV-8: polypropylene glycol di(meta) acrylate

(Number average molecular weight 200~1,000)

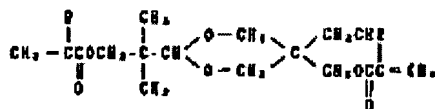
IV-9: neo pentyl glycol di(meta) acrylate.  
 IV-10: 1,3 butane diole di(meta) acrylate  
 IV-11: 1,4 butane diole di(meta) acrylate  
 IV-12: 1,6-hexane diole di(meta) acrylate  
 IV-13: hydroxy pivalic acid ester neopentyl glycol di(meta)

#### Acrylate



IV-14: (但し、RはH又はCH<sub>3</sub>、)

(However, R is H or CH<sub>2</sub>)



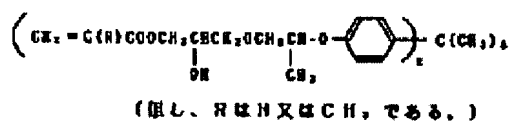
IV-15:

(However, R is H or CH<sub>2</sub>)

IV-16: bisphenol A di(Meta) acrylate

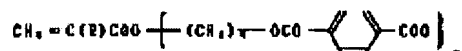
IV-22: dipenta erythrytol monohydroxy penta (meta) acrylate

Furthermore, as the specific examples other than above described, there are neoma MA-305 (IV-23), neoma -BA-60 (IV-24), neoma-TA-505 (IV-25), neoma-TA-401 (IV-26), neoma PHA405X (IV-27), neoma TA705X (IV-28), neoma EA400X (IV-29), Neoma EE401X (IV-30), Neoma EP405X (IV-31), Neoma HB601X (IV-32), Neoma HB605X (IV-33) (made by Sanyo Kasei Co); YAYARD NY-220 (IV-34), HX-620 (IV-35), D-310 (IV-36), D-320 (IV-37), D-330 (IV-38), DPHA (IV-39), DPCA-20 (IV-40), DPCA-30 (IV-41), DPCA-60 (IV-42), DPCA-120 (IV-43) (made by Nippon Kasei Co); IV-44: FA-713A made by Hitachi Kasei Co IV-45:



(However, R is H or CH<sub>2</sub>)

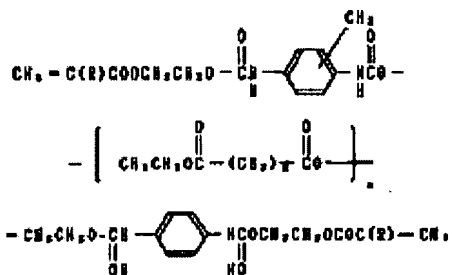
IV-46:



(n, x are integers of 1~10)

(However, R is H or CH<sub>2</sub>)

IV-47:



(n, x are integers of 1~10)

(However R is H or CH<sub>2</sub>)

IV-48: polyethylene glycol 400 di(meta) acrylate

IV-49: 1,3-bis(3'-acryloxyethoxy-2'-hydroxypropyl)

5,5-dimethylhydantoin

IV-50:



(However, R is H or CH<sub>2</sub>)

IV-51:



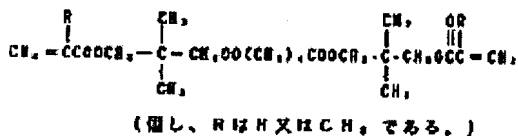
(However, R is H or CH<sub>2</sub>)

IV-52:



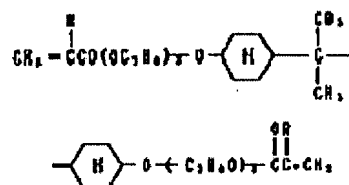
(However, R is H or CH<sub>2</sub>)

IV-53:



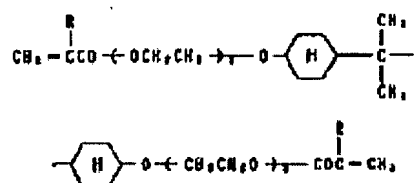
(However, R is H or CH<sub>2</sub>)

IV-54:



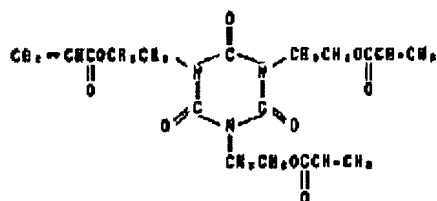
(However, R is H or CH<sub>2</sub>)

IV-55:

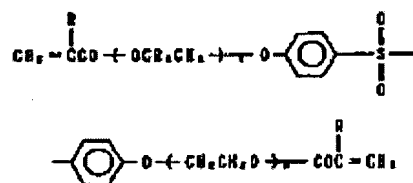


(However, R is H or CH<sub>2</sub>)

IV-56:

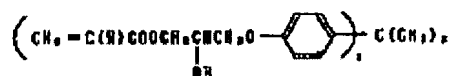


IV-57:



(However, R is H or CH<sub>2</sub>)

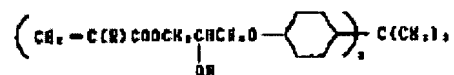
IV-58:



First one

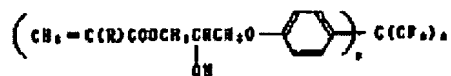
(However, R is H or CH<sub>2</sub>)

IV-59:



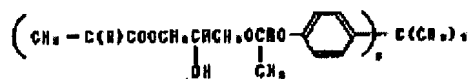
(However, R is H or CH<sub>2</sub>)

IV-60:



(However, R is H or CH<sub>2</sub>)

IV-61:



(However, R is H or CH<sub>2</sub>)

IV-62:



First one

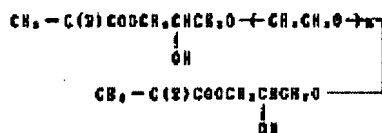
(However, R is H or CH<sub>2</sub>)

IV-63:



(However, R is H or CH<sub>2</sub>)

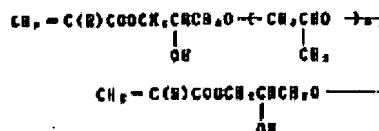
IV-64:



(m is an integer of 1~10)

(However, R is H or CH<sub>2</sub>)

IV-65:

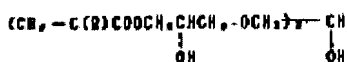


First one

(m is an integer of 1~10)

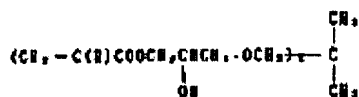
(However, R is H or CH<sub>2</sub>)

IV-66:



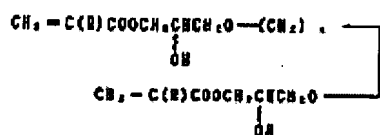
(However, R is H or CH<sub>2</sub>)

IV-67:



(However, R is H or CH<sub>2</sub>)

IV-68:



First one:

("l-el" is an integer of 2~9)

IV-69:



(However, R is H or CH<sub>2</sub>)

IV-70:



(However, R is H or CH<sub>2</sub>)

IV-71:



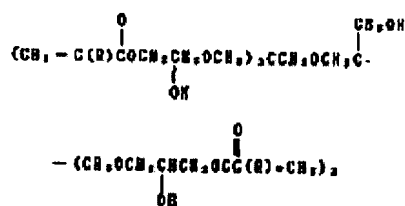
(However, R is H or CH<sub>2</sub>)

IV-72:



(However, R is H or CH<sub>2</sub>)

IV-73:



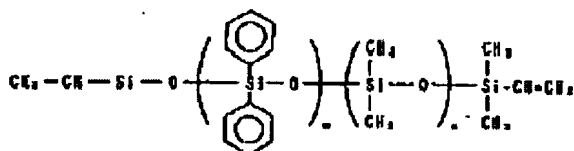
(However, R is H or CH<sub>2</sub>)

IV-74:

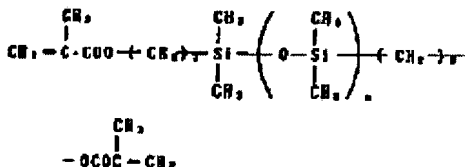


(However, R is H or CH<sub>2</sub>)

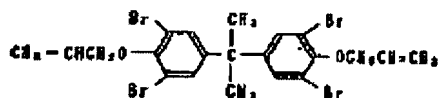
IV-75:



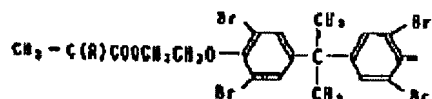
IV-76:



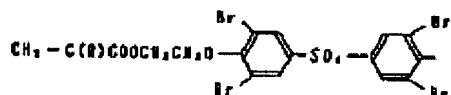
IV-77:



IV-78:



See first one



IV-79:

See second one

(However, R is H or CH<sub>2</sub>)

Multifunctional acrylate is important in imparting the curable characteristics, flexibility, mechanical strength and the like of insulation material of the present invention.

It is very important that the insulation material of radial polymerizable system of the present invention contains

polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer as the essential ingredient.

The insulation material of radical polymerizable system of the present invention is constituted by polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer and alpha, beta - unsaturated ethylene monomer, however, considering the absolute value of insulation performance, it is preferred that it is constituted by polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer and polyfluoroalkyl group - containing poly alpha, beta - unsaturated ethylene monomer as essential ingredients.

Regarding the synthesis ratio of polyfluoroalkyl group - containing monoacrylate (1-a), polyfluoroalkyl group - containing multi functional acrylate (1-b), alpha, beta - unsaturated ethylene dicarboxylic acid or its ester (II), monoacrylate without polyfluoroalkyl group (III), multifunctional acrylate without polyfluoroalkyl group (IV) in the insulation material of radical polymerizable system of the present invention, it can be optionally selected by matching with the mechanical properties after curing, however, considering curing characteristics, the flexibility of the insulation body after curing, and insulation performance of the present invention, its weight ratio is as follows,

$$(1-a) + (1-b) + (II) : (III) + (IV) = 3 : 100 \sim 100 : 0$$

Here,  $(1-a) + (1-b) : (II) = 50 : 50 \sim 100 : 0$

(III): (IV)=0: 100~100:0 and also

(I-a): (1-b)= 1:99~100:0

And, from insulation performance stand point, it is preferred that as polyfluoroalkyl group - containing monofunctional monomer, fluorine containing monoacrylate (1-a), polyfluoroalkyl containing alpha, beta - unsaturated ethylene dicarboxyl acid contains its ester.

The insulation material of the present invention is polymerized and cured by providing light, electron beams, radiation energy, furthermore, heat, and can form desired insulation body. As the polymerization starting energy, in case light such as UV beams are used, desired light polymerization starting agent well known in the industry can be used. As such light starting agent, there are for instance, V-1: benzophenone, V-2: acetophenone, V-3: benzoine, V-4: benzoine ethyl ether, V-5: benzoine isobutyl ether, V-6: benzyl methyl ketal, V-7: azo bisbutylo nitrile, V-8: hydroxy cyclohexyl phenyl ketone, v-9: 2-hydroxy -2- methyl -1- phenyl propane -1- on and the like, and depending on the needs, light sensitizer such as amine compound, or phosphorous compound and the like are added, thus speeding the polymerization. From the stand point of accelerating polymerization curing and /or improving the heat resistance and light fastness of insulation body, monofunctional or multi functional thiol group - containing compound can be added. In case it is polymerized and cured by electron beams or radiation beams, it is not particularly

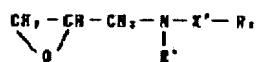
necessary to add polymerization starting agent and the like.

And, in case heat is used as polymerization starting agent, in the presence of the non-catalyst or the polymerization starting agent such as azo bis isobutylo nitrile, benzoyl peroxide, methyl ethyl ketone peroxide - naphthenic acid cobalt and the like, for instance, it can be polymerized and cured at 80~200 deg C.

According to the opinion of the inventors herein, from the stand point of workability in forming insulation body, productivity, and economic aspect, and the performance after completing the insulation body, compared with curing by heat, it is preferred that any one of UV beams, electron beams, and radiation beams be used for curing. Among them, the method to polymerized and cured by UV beams is the simplest and also most economical.

On the other hand, the cation polymerization system among the insulation material of the present invention is constituted by polyfluoroalkyl group - containing monoepoxy monomer mainly, hence, depending on needs, monoepoxy monomer or polyepoxy monomer without polyfluoroalkyl group is used.

As polyfluoroalkyl group - containing monoepoxy monomer (IV), for instance,



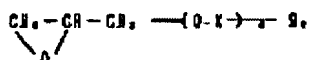
(in the formula, R<sub>1</sub> is same as before, X' is -SO<sub>2</sub>-, -CO-,  
-(CH<sub>2</sub>)<sub>2</sub> (however, n' is 1~10), or -CH<sub>2</sub> CK CK?

|

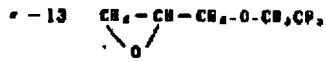
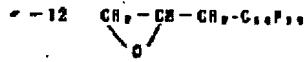
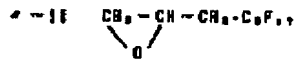
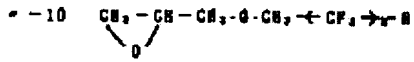
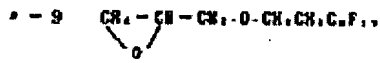
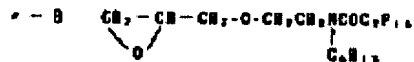
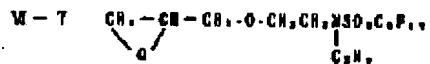
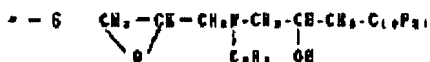
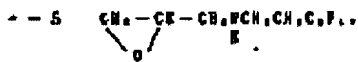
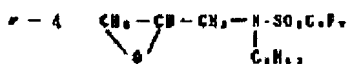
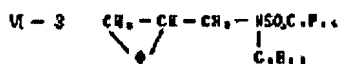
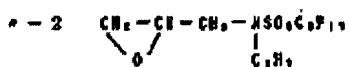
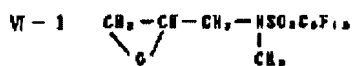
OR

R1 is H or an alkyl group with carbon number 1-6)

Furthermore, a compound represented by the following general formula can be listed,



(In the formula, Rf?, X are same as before, d is zero (0) or one (1)). The following can be listed as more specific compounds than those.



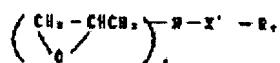
As the polymerizable monomer that can be combined with above

described polyfluoroalkyl group - containing monomer, for instance, polyepoxy monomer can be listed.

In the present invention, the polyepoxy monomer refers to the monomer which contains at least two units of polymerizable epoxy groups.

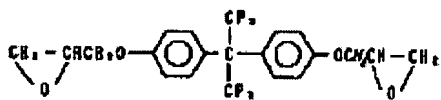
Polyepoxy monomers can be largely classified into polyfluoroalkyl group or polyfluoroalkylene group - containing polyepoxy monomer (VI) and polyepoxy monomer without polyfluoroalkyl group or polyfluoroalkylene group (IV).

As polyfluoroalkyl group or polyfluoroalkylene group - containing polyepoxy monomer (VI), the following can be listed: the compound wherein the hydrogen atoms in the structure ingredient which excludes the glycidyl group in polyepoxy monomers without polyfluoroalkyl group or polyfluoroalkylene group later described is substituted by fluorine atoms or perfluoro alkyl group with carbon number of 1~20 or fluoro aliphatic group, Or the compounds represented by following general formula

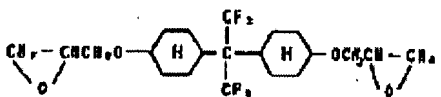


(in the formula, R1, X' are same as before),

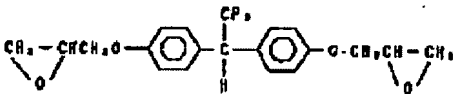
Furthermore, ones described in USP 3,720, 639, USP 3,873, 430, USP 4,132, 681, USP 4,284, 747, USP 4,356, 296 and the like. As more detailed specific examples of these, for instance, the following compounds can be listed



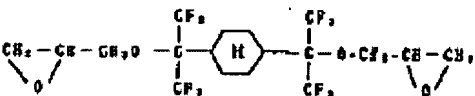
2



3

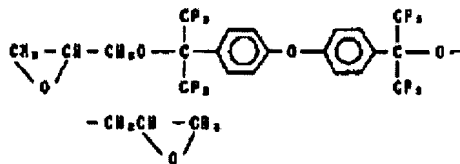


4

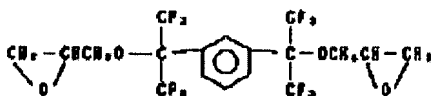


(P, q are integers of 1~10)

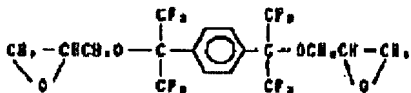
VI - 5



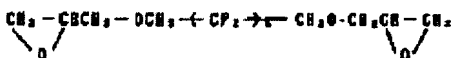
6



7



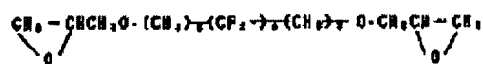
8



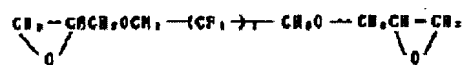
First one

(p, q are integers of 1~10)

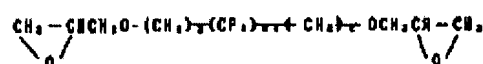
VI-9



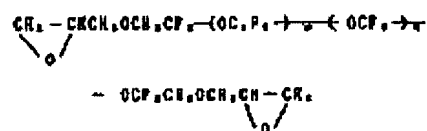
- 10



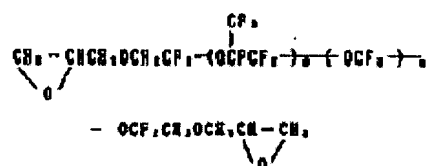
VI-11



- 12

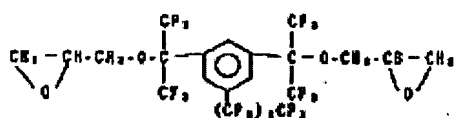


VI-13

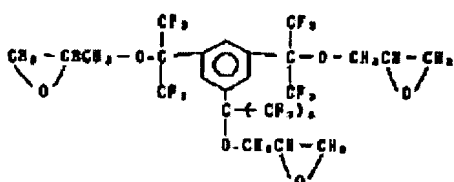


(p, q は 1 ~ 10 の整数である。)

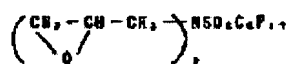
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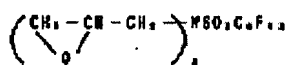
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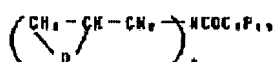
W-16



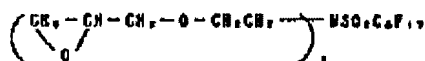
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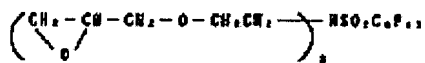
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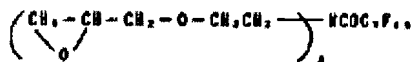
W-19



W-20



W-21

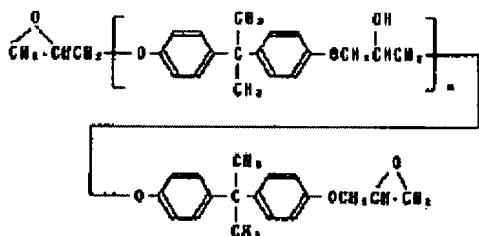


And, the present invention is not limited to the above described special example, of course.

As the cation polymerizable ingredient of insulation material of the present invention, other than above described each epoxy monomer group, monoepoxy monomer (VI) without polyfluoroalkyl group or polyfluoroalkylene group and polyepoxy monomer (IX) without polyfluoroalkyl group or polyfluoroalkylene group is preferably used.

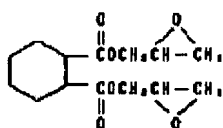
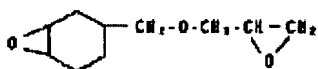
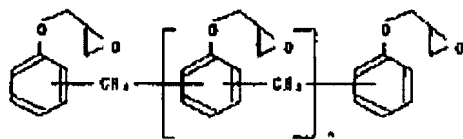
the monoepoxy monomer (VI) without polyfluoroalkyl group are glycidyl ether compound of monohydroxy compound and glycidyl esterified product of monocarboxylic acid compound, for instance, there are olefin oxide, octylene oxide, butyl glycidyl ether, glycidyl methacrylate, acryl glycidyl ether, styrene oxide, phenyl glycidyl diether, n-butyl phenol glycidyl ether, 3-pental decil

Furthermore, silicone chain containing epoxy compound as in (n=1~150)

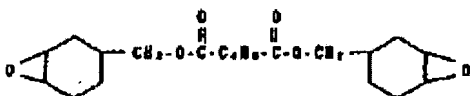
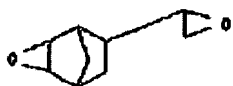
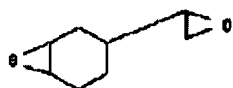
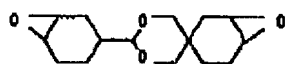
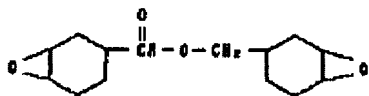

$$\text{CH}_3 - \text{CH} - \text{CH}_2 - \text{O} - (\text{CH}_2)_2 - \text{O} - \text{Si}(\text{CH}_3)_2 - \text{O} - \text{Si}(\text{CH}_3)_2 - \text{CH}_2 - \text{CH} - \text{CH}_3$$
$$\begin{array}{c} \text{Br} \\ | \\ \text{Br}-\text{C}_6\text{H}_2-\text{Br} \\ | \\ \text{Br} \end{array} \text{OCN, CH}_2\text{NCH}_2 \sim \text{CH} \sim \text{CH}_2 \text{O} \text{O} \text{O}$$

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type epoxy resin, it is represented by the general formula,



(In the formula,  $n$  is 0-20). More specifically, epicoat 152, 154, 172 (made by Shell Kagaku Co), Ararudaito EPN (made by Chiba Gaigi Co), SER 431, 438 and 439 (made by Dow Chemical Co) and the like can be listed, as the lipid ring type epoxy resin, for instance

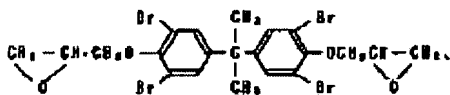


Can be listed, as commercial product,  
Araildiato CY-175, -176, -179, -182, -184, 192 (made by Chiga Gaigi

Co), Chisonox 090, 091, 092, 301, 313 (made by Chisso Co), CYRACURE 6100, 6110, 6200, ERL 4090, 4617, 2256, 5411 (made by Union Carbide Co) and the like,

As the polyhydric glycidyl ether derived from aromatic polyhydric alcohol, there are, for instance, diglycidil ether of 1~16 mol additional mass of alkylene oxide of bisphenyl A, diglycidil ether of 1~16 mol additional mass of alkylene oxide of bisphenyl F, Diglycidil ether of 1~16 mol additional mass of alkylene oxide of bisphenyl S and the like,

as the polyhydric glycidyl ether type of aliphatic polyhydric alcohol, there are, for instance, ethylene glycol diglycidil ether, polyethylene glycol diglycidil ether, propylene glycol diglycidil ether, polypropylene glycol diglycidil ether, neo pentyl glycol diglycidil ether, 1,6- hexane diol diglycidil ether, glycerin diglycidil ether, trimethylol propane triglycidyl ether, diglycidil ether of hydrogen added bisphenol A, and 2,2 dipromoneo pentyl glycol diglycidil ether and the like, furthermore, hetero cycle type glycidyl amine resin, resin derived from triglycidil -p- amino phenol, resin which has triazine as a base, and hydantoin epoxy resin, and silicon chain containing diepoxy monomer like the following general formula,


$$\text{CH}_3-\text{CH}-\text{CH}_2\text{O}-\text{C}_6\text{H}_2(\text{Br})_2-\text{SO}_2-\text{C}_6\text{H}_2(\text{Br})_2-\text{OCH}_2\text{CH}-\text{CH}_3$$

Regarding the insulation material of cation polymerization system of the present invention, it is important that it contains polyfluoroalkyl group - containing monoepoxy monomer as the essential ingredient.

Polyepoxy monomer is important in order to impart the insulation performance, curability, flexibility of the insulation body after

curing, mechanical strength and the like of the insulation material of the present invention. From the stand point of insulation performance, it is preferred, as described before, that 3 weight% or more of fluorine atoms are contained in the insulation material of the present invention, however, giving consideration to the curability, flexibility of insulation material after curing, and mechanical strength, the composition ratio of polyfluoro alkyl group - containing monoepoxy monomer(VI), polyfluoroalkyl group or polyfluoroalkylene group - containing polyepoxy monomer (VI), monoepoxy monomer without polyfluoroalkyl group or polyfluoroalkylene group (VII), polyepoxy monomer (IV) without polyfluoroalkyl group or polyfluoroalkylene group occupied in insulation material is in weight ratio:

$$(VIII) + (VI) : (VX) + (VII) = 0:100 \sim 97:3$$

Here, the followings are preferred

$$(VII)+(VX) : (VI) + (VII) = 0:100 \sim 97:3$$

And

$$(VIII) : (VX) = 0:100 \sim 100:0$$

$$(VI) : (VII) = 1:99 \sim 100:0$$

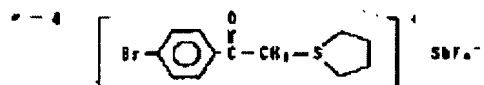
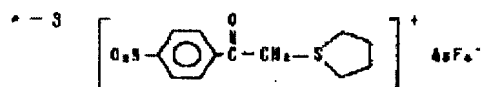
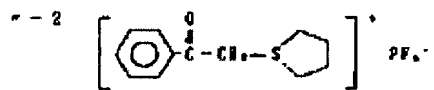
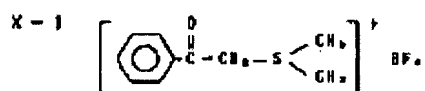
Regarding the insulation material of cation polymerization system of the present invention, curing agents are combined such as boron monoethylene amine trifluoride, boron benzyl amine trifluoride, boron acid chlorine tetra fluoride, dibasic acid hydrazine such as dihydrazine adipate; alkyl boron such as triethyl

amin boron and the like, Phenylene diamine - zinc bromide complex, dicyan diamide, p, p' -diamine diphenyl methane, p, p?' -diamino diphenyl sulfonic acid, 2-ethyl - 4 methyl imidazole and the like, and cured in the room temperature or while heated, thus desired insulation body is formed, however, according to the knowledge of the present invention, the method to polymerize and cure by using the light such as UV beams in the presence of light polymerization starting agent, compared with the curing by heat, is preferred from the standpoint of productivity, and economy and the like.

As favorable light polymerization starting agent at the time, following can be listed:

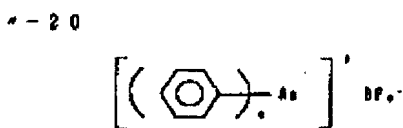
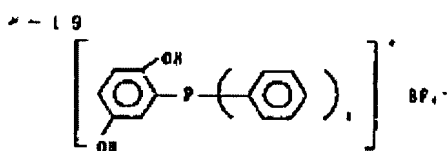
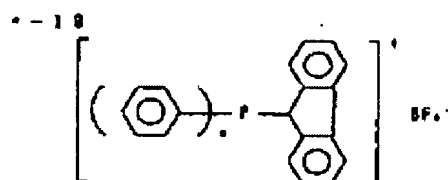
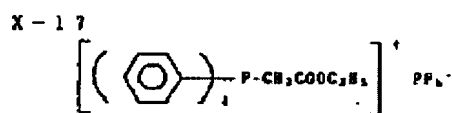
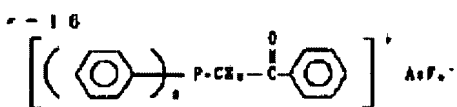
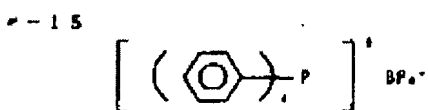
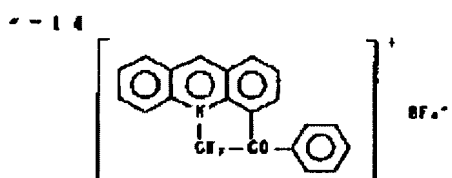
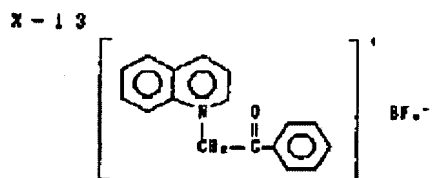
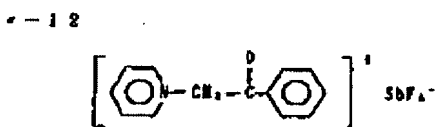
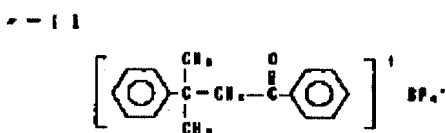
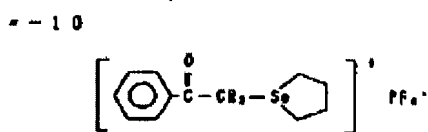
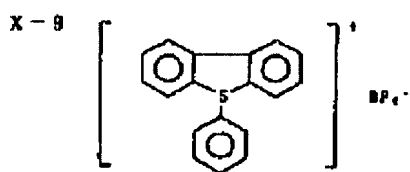
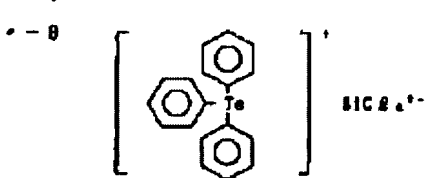
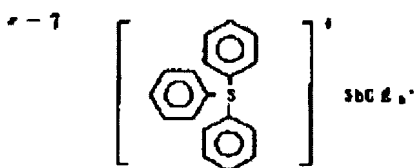
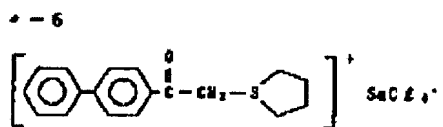
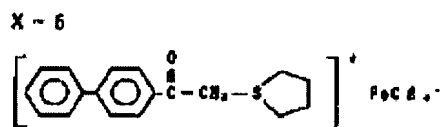
One is aromatic group, onium chloride having light sensitivity which contains the elements that belongs to VIa type in the periodic table VIa? Described in Japan Patent Disclosure Sho 52-14278, another one is aromatic onium chloride which contains the elements that belong to the Va type described in Japan Patent Disclosure Sho52-14279.

Regarding the aromatic onium chloride that contains the elements that belong to VIa type or Va type, they are represented by the following general formula



[in the formula, R1? is monovalent organic aromatic group, R10? is monovalent organic aliphatic group selected from alkyl group, dichloro alkyl group, and substituted alkyl group, R11 is a polyhydric organic group which constitutes the chlorine ring or condensed ring structure selected from aliphatic group or aromatic group, D is an elements that belong to VIa group selected from sulfur, selenium, or tellurium or, Va group selected from nitrogen, phosphorus, arsenic, antimonies and bismus; M stands for metal or metalloid, Q stands for halogen group respectively; a is an integer of 0~3 in case D is an element that belongs to VIa group however, it is an integer of 0~4 in case D is an element that belongs to V2 type; b is an integer of 0~2; c is an integer of 0 or 1 in case it is an element that belongs to mega VIa group and is an integer 0~2 in case D is an element that belongs to Va group; f is an integer of 2~7 with valence of M and; e is an integer larger than 1 but 3 or less and also, the sum of a, b, and c, in case of the element that belongs to mega VIa group, is 3 (the valence of X), and in case D is the

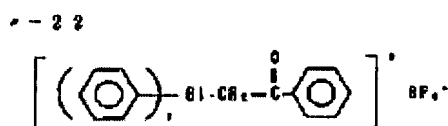
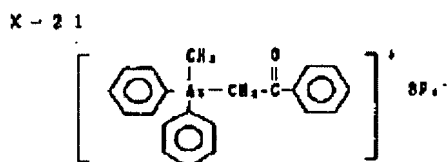
element that belongs to Va group, is 4 (valence of X),  $d=e-f$ ], the characteristics is that when light hits these, onium salt emits Lewis acid, and this starts the polymerization of epoxy resin. As these special examples, the following compound can be listed.



In

addition to those, The aryl diazonium compound described in Japan Patent Disclosure Sho63-8153 and diaryl iodonium chloride compound

described in J. V. Grivello and J. H. W. Lam, Macromolecules ID (6), page 1307~1315 (1977), and the iron - allene complex type compound as shown in the chemical structure



Those compounds which generated Lewis acid by lights can be favorably used as the light polymerization starting agent of the epoxy resin of the present invention.

Regarding the ratio of the light polymerization starting agent occupied in insulation material of the present invention, Regardless of the radical polymerization system or cation polymerization system, it is 0.01~20 weight parts per 100 weight parts of insulation material of the present invention, more preferably 0.1~10 weight parts. If the usage ratio of the light polymerization starting agent is within above described range, it sufficiently accelerates the curing speed, hence, the productivity of insulation body becomes good, and as in case more than necessary amount of light polymerization starting agent is used, it is difficult to adversely impact on the mechanical strength.

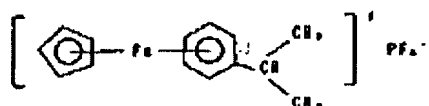
Furthermore, regarding the insulation materials of the present invention, from the standpoint of adhesiveness with the base to be

insulated, and also, efficiency of workability and stability when coated on the insulation body and the like, coupling agent, anti-foaming agent, leveling agent, coloring agent, surface active agent, plastic agent, surface improving agent, and viscosity adjustment agent, and from standpoint of improving the heat resistance, anti-oxidant agent, inflammation retarding agent, and from the standpoint of improving the light fastness, light fastness stabilizer and the like can be introduced, depending on needs.

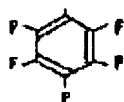
As the viscosity adjustment agent, macromolecular compound can be listed. Among them, synthetic resin is particularly preferred, for instance, synthetic resins such as acrylic type resin, epoxy type resin, silicon type resin, fluorine type resin and the like ~~form ... light film~~ Can be listed, anyone of them can be used if it meets the above described conditions. Regarding synthetic resin, any one of thermo plastic resin, thermo curable resin, and active energy beam curing resin can be used of course.

As the specific examples of synthetic resin, one obtained by radical polymerization mechanism or cation polymerization synthesis mechanism can be listed. One example of former one is polymers of alpha, beta - unsaturated ethylene monomer, one example of the latter case is the polymer of epoxy monomer, furthermore, vinyl containing fluoro polysiloxane , hydrogen atom containing fluoro silicone (hereafter EP208239), fluoro alkyl methacrylate/ ortho methyl phenyl maleic imide copolymer (Japan Patent Disclosure Sho 60-235817),

chloro trifluoro ethylene/ 2,2,3,3- tetrafluoro propyl vinyl ether/  
hydroxyl group - containing vinyl ether copolymer (EP121934), fluoro  
polyimide (Journal of Photopolymer Science and Technology. I (1)  
120.121 (1988)), silicon coating material (Toshiba review, 38 (2),  
171-174 (1983)), poly(tetra fluoro ethylene), poly(fluoro  
vinylidene), poly(vinylidene fluoride/ tetrafluoro ethylene),  
poly(tetra fluoro ethylene/hexafluoro propylene), poly(tetra fluoro  
ethylene/ propylene), poly(ethylene /tetrafluoro ethylene),  
poly(vinylidene fluoride/hexa fluoro propylene), poly(monofluoro  
ethylene), poly(trifluoro ethylene), poly(vinyl ether fluoride),  
poly(tetrafluoro ethylene / vinyl ether fluoride), poly(perfluoro  
styrene), saitop <sup>®</sup> (made by Asahi Glass Co) or Teflon <sup>®</sup> AF (made  
by DuPon) that contain macro molecules and fluorine atoms in the  
main chain, and it can be, for instance, polymer of fluoro epoxy  
compound as in the chemical structure



(However, R1 is a fluoro alkyl group with carbon number of 1~20),  
or the polymer with  $\text{-(CH}_2\text{ CH)-}_n$



As the synthetic resin, particularly fluorine containing  
polymer (X1) is preferred. As fluorine containing polymer (x1),  
ones obtained by radical polymerization mechanism or cation

polymerization mechanism can be listed, as one example of the former, the polymer obtained using polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer as the essential ingredient, and the as one example of the latter, the polymer obtained using polyfluoro alkylene group - containing epoxy monomer as the essential ingredient can be listed, however, in order to avoid the confusion, the following explanation will be done in detail regarding the former.

According to the knowledge of the inventors herein, from the stand point of aforementioned polyfluoroalkyl group - containing mono alpha, beta - unsaturated ethylene monomer, particularly, fluorine containing acrylate (I), as synthetic resin, it is preferred that polyfluoroalkyl group - containing alpha, beta - unsaturated ethylene monocarboxylic acid, or its ester compound, and/or polyfluoroalkyl group - containing alpha, beta - unsaturated ethylene dicarboxylic acid or its ester compound polymer (X1->A) is preferred.

As the polyfluoroalkyl group - containing alpha, beta - unsaturated ethylene monocarboxylic acid or its ester compound, for instance, aforementioned fluorine containing monoacrylate (I-A) can be listed.

As fluorine containing alpha, beta - unsaturated ethylene dicarboxylic acid or its ester compound, for instance, among the aforementioned alpha, beta - unsaturated ethylene ester carboxylate (II), polyfluoroalkyl group - containing one can be listed.

And, of course, the present invention is not limited by the above described specific examples.

According to the knowledge of inventors herein, from the standpoint of compatibility with fluorine containing acrylate (I), and the ease to obtain raw materials and economy, as the monomer which constitutes fluorine containing polymer (XI-A) of the present invention, fluorine containing monoacrylate (I-a) is particularly preferred. Of course, there is no problem if fluorine containing monoacrylate (I-a) and polyfluoro alkyl group - containing alpha, beta- unsaturated ethylene dicarboxylic acid or ester compound are combined.

As the monomer that constitutes fluorine containing polymer (XI-A), other than fluorine containing monoacrylate (I-a), and polyfluoro alkyl group - containing alpha, beta- unsaturated ethylene dicarboxylic acid or ester compound, in order to improve the compatibility with fluorine containing acrylate (I) of fluorine containing polymer (XI-A) or to adjust the mechanical strength and flexibility of insulation body obtained by curing the insulation material of the present invention, mono alpha, beta - unsaturated ethylene monomer without polyfluoroalkyl group in molecules can be introduced.

As the mono alpha, beta - unsaturated monomer without polyfluoroalkyl group, other than acrylic acid, methacrylic acid, there are monoacrylate without polyfluoro alkyl group, alpha, beta

- unsaturated ethylene dicarboxylic acid without polyfluoroalkyl group, and its ester compound.

As the monoacrylate without polyfluoroalkyl group, listed in the aforementioned monoacrylate (II), as alpha, beta - unsaturated ethylene dicarboxylic acid without polyfluoroalkyl group or its ester compound, the compound represented by

(In the formula, R12 and R13 are H, Cl, CN or CH2, and R12 and R13 can be same or different), or the compound without polyfluoroalkyl without polyfluoroalkyl group in the aforementioned alpha, beta - unsaturated ethylene dicarboxylic acid or its ester (II) can be listed.

As mono alpha, beta - unsaturated ethylene monomer without polyfluoroalkyl group other than monoacrylate without polyfluoroalkyl group and alpha, beta - unsaturated ethylene dicarboxylic acid without polyfluoroalkyl group and its ester compound, aforementioned diluted monomer can be used. Of course, the present invention is not limited to above described specific examples.

As the fluorine containing polymer of the present invention (XI), the polymerizable alpha, beta - unsaturated ethylene group - containing fluorine containing resin can be preferably used wherein functional group - containing alpha, beta - unsaturated ethylene monomer is introduced into the functional group which exist on the side chain and /or at the end of aforementioned fluorine polymer

(XI-A) directly or via combined group, thus producing said fluorine containing resin.

Regarding fluorine containing polymer (XI-A), as the copolymerizable monomer which can provides functional group on side chain, there are aforementioned epoxy group - containing monomer, hydroxyl group - containing monomer, carboxyl group - containing monomer, isocyanate group - containing monomer and the like in various alpha, beta-unsaturated monomers. And the functional group at the end of fluorine containing polymers (XI-A) can be introduced by executing the normal radical polymerization reaction in the co-presence of mercapto group - containing alcohol or mercapto group - containing carboxylic acid called chain transfer agent.

As the functional group - containing alpha, beta - unsaturated ethylene monomer which can react with the functional group that exists at the side chain and/or end in fluorine containing polymer (XI-A), there are epoxy group - containing monomer, hydroxyl group - containing monomer, carboxyl group - containing monomer, isocyanate group - containing monomer and the like.

The polymerizable alpha, beta - unsaturated ethylene group - containing fluorine resin (XI-B) of the present invention can be synthesized by following method.

1). in case the functional group of the side chain and/or ends of fluorine containing polymer (XI-A) is epoxy group and/or hydroxyl group, a method is such that said functional group and carboxyl group

- containing alpha, beta - unsaturated ethylene monomer are reacted, and alpha, beta - unsaturated ethylene group is introduced.

2). in case the functional group of the side chain and/or ends of fluorine containing polymer (XI-A) is carboxyl group, a method is such that said functional group and epoxy group - containing alpha, beta - unsaturated ethylene monomer and/or hydroxyl group - containing alpha, beta - unsaturated ethylene monomer are reacted by esterification, thus alpha, beta - unsaturated ethylene group is introduced.

3). In case the functional group of the side chain and/or ends of fluorine containing polymer (XI-A) is a hydroxyl group, a method is such that said functional group and hydroxyl group - containing alpha, beta - unsaturated ethylene monomer are reacted by adding urethane via polyisocyanate compound, thus, alpha, beta - unsaturated ethylene group is introduced, or a method is such that said functional group and isocyanate group - containing alpha, beta - unsaturated ethylene monomer are reacted by adding urethane, thus alpha, beta - unsaturated ethylene group is introduced.

4). in case the functional group of the side chain and/or ends of fluorine containing polymer (XI-A) is an isocyanate group, said functional group and hydroxyl group - containing alpha, beta - unsaturated ethylene monomer are reacted by adding urethane, thus alpha, beta - unsaturated ethylene group is introduced.

Here, as the carboxyl group - containing alpha, beta -

unsaturated ethylene monomer, there are, for instance, acrylic acid, methacrylic acid, 2-acryloyl oxy ethyl succinic acid, 2-methacryloyl oxy ethyl succinic acid, 2-acryloyl oxy ethyl maleic acid, 2-methacryloyl oxy ethyl maleic acid, 2-acryloyl oxy ethyl phthalic acid, 2-methacryloyl oxy ethyl phthalic acid, 2-acryloyl oxy ethyl hexahydro phthalic acid, 2-methacryloyl oxy ethyl hexahydro phthalic acid, beta-carboxy ethyl acrylate, beta-carboxy ethyl methacrylate and the like; as epoxy group - containing alpha, beta - unsaturated ethylene monomer, for instance, glycidyl acrylate, glycidyl methacrylate, meta glycidyl acrylate, meta glycidyl methacrylate and the like; as hydroxyl group - containing vinyl monomer, for instance, hydroxy ethyl acrylate, hydroxy ethyl methacrylate, hydroxy butyl acrylate, and hydroxy butyl methacrylate and the like that are dialcohol compound, and monoester compounds with acrylic acid or meta acrylic acid, capro lacton denatured acrylate or methacrylate and the like; as isocyanate group - containing vinyl monomer, urethane bond containing compound synthesized from for instance, 2-isocyanate ethyl acrylate, or methacrylate, and hydroxyl group - containing vinyl monomer and polyisocyanate compound; and as polyisocyanate compound, they are compound having two units or more of isocyanate groups, such as, for instance, trilen diisocyanate, 4,4'-diphenyl methane diisocyanate, xylen diisocyanate, hexa methylene diisocyanate, lysine diisocyanate, 4,4' - methylene bis(cyclo hexyl isocyanate), methyl cyclohexane

-2,4-diisocyanate, methyl cyclohexane -2,6- diisocyanate, 1,3-(isocyanate methyl) cyclohexane, isophorone diisocyanate, trimethyl hexa methylene diisocyanate, dianisidine diisocyanate, phenyl diisocyanate, halogenated phenyl diisocyanate, methylene diisocyanate, butylene diisocyanate, propylene diisocyanate, octadecilen diisocyanate, 1,5-naphthalene diisocyanate, triphenyl methane triisocyanate, naphthalene diisocyanate and the like, among these polyisocyanate, trilene diisocyanate, isophorone isocyanate, and lysine diisocyanate are preferred.

And, regarding the detailed specific examples and synthesis method of polymerizable unsaturated bonded group - containing fluorine resin of the present invention is described in, for instance, GB 2199332.

In the curable insulation material of the present invention, regarding the fluorine containing polymer (XI) used when needed, when curable insulation material of the present invention is coated with high speed on electrical wire conductive body, it is important that no coating defect such as pin holes and the like is generated, leveling characteristics, viscosity and coating suitability which are sufficient to coat on the base material are met, and suitable viscosity is preferred. Particularly, as the coating material for thin film insulation electrical wire, it is important that there is no defect such as pin holes, and it has fast speed productivity, and is economical when electrical wire conductive bodies are coated.

The fluorine content amount in fluorine containing polymer (XI) of the present invention is not limited, however, from the standpoint of compatibility with fluorine containing acrylate (I), 0.2 weight% or more is preferred, and particularly, 1.0 weight% or more is preferred. Furthermore, fluorine containing polymer (XI), if it contains aforementioned fluorine containing monoacrylate (I-a) as its constituent unit, the compatibility with fluorine containing acrylate (I) improves, furthermore, leveling characteristics and adhesives with the base material improve, as the fluorine monomer which constitutes fluorine containing polymer (XI), fluorine containing monoacrylate (I-a) is preferred.

The composition ratio of fluorine containing polymer (XI), the total (I) of fluorine - containing monoacrylate and fluorine - containing multi functional acrylate, monoacrylate without polyfluoroalkyl group (II), and multiple functional acrylate without polyfluoroalkyl group (IV) can be optionally selected by matching with desired viscosity, mechanical strength after curing and the like, however, from the standpoint of curing characteristics, the flexibility of insulation material after curing, and the high degree of insulation performance which is the purpose of the present invention, the following weight ratio is preferred:

$$(XI): (I) + (II) + (IV)$$

$$= 0.01: 99.99 \sim 10,000: 1$$

$$(XI) + (I): (II) + (IV)$$

$$= 1.99 \sim 100:0$$

$$(XI): (I) = 0.01: 99.99 \sim 10,000: 1$$

Fluorine containing polymer of the present invention is produced by the well known polymerization method in this industry, for instance, methods such as radical polymerization anion polymerization and the like, using heat, light, electron beams, and radiation beams and the like as the polymerization starting energy, however, industrially, radical polymerization that uses heat and/or light as polymerization starting energy is preferred. As the polymerization form by these polymerization methods, any one of solid polymerization, bath polymerization, and emulsification polymerization can be adopted. In case heat is used for polymerization starting energy, non catalyst or azo bisisobutyronitrile, benzoyl peroxide, t-butyl peroxy - 2- ethyl hexanoate, methyl ethyl ketone peroxide - naphthenic acid cobalt and the like that are polymerization starting agents or in case light such as UV beams is used, well known desired light polymerization starting agents in said industry (for instance, the compound and the like shown in aforementioned V-1~11), and depending on the needs, or light sensitizer such as amine compound or phosphoric compound and the like can be added, thus accelerating the polymerization. And in these radical polymerization, depending on needs, lauryl mercaptan, thio glycolic acid octyl, gamma-mercapto propyl trimethoxy silane, C<sub>6</sub>F<sub>17</sub>CH<sub>2</sub>CH<sub>2</sub>SH and the like that are mercapto group - containing

transfer agents can be combined, thereby, thus adjusting the polymerization degree of fluorine containing polymer (I). In case electron beams or radiation beams are used for polymerization, it is not necessary to add polymerization starting agent particularly. And in case fluorine containing polymer (XI) is obtained by bath polymerization, as solvent, as long as it does not adversely impact the polymerization reaction, there is no limitation in usage.

Regarding the insulation material of the present invention, in case fluorine type monomer which is the constituent monomer of fluorine containing polymer (XI) and non fluorine type monomer is equivalent to fluorine containing monoacrylate (I-a) or monoacrylate without polyfluoroalkyl group (II), the polymerization of fluorine containing polymer (XI) is stopped at the stage where unreacted fluorine type monomer and non fluorine type monomer remain, and after that, depending on needs, specified amount of fluorine containing multifunctional acrylate (I-b) and/or multifunctional acrylate without polyfluoroalkyl group (IV) is added and mixed, the resin composition of the present invention can be obtained easily.

As coupling agents, there are, for instance, silane type, titanium type, zircon- aluminate type; among them, dimethyl dimethoxy silane, dimethyl diethoxy silane, methyl trimethoxy silane, dimethyl vinyl methoxy silane, phenyl trimethoxy silane, gamma-chloro propyl trimethoxy silane, gamma-chloro propyl methyl dimethoxy silane, gamma-amino propyl triethoxy silane, gamma-mercapto propyl triethoxy

silane, gamma-glycidoxy propyl trimethoxy silane, gamma-glycidoxy propyl methyl dimethoxy silane, gamma-methacryloxy propyl methoxy silane, gamma-methacryloxy propyl methyl dimethoxy silane, gamma-acryloxy propyl methyl trimethoxy silane, gamma-acryloxy propyl methyl dimethoxy silane and the like that are silane type is particularly preferred.

As the antifoaming agent, leveling agent, surface active agent, surface improving agent to be used, and fluorine type is preferred.

As antioxidant agent, there are, for instance, ion type, phosphorus type, hyndard phenol type and the like, however, among them, one with thiol group or hyndard phenol type is particularly preferred.

As fire retarding agent, there are, for instance, fire retarding agent of bromo type, zinc compound, antimony type compound, phosphorus type compound or one with these two types or more combined.

As bromo type fire retarding agent, there are deca bromo dipheyl oxide, hexa bromo benzene, hexybromo cyclododecan, dodeca chloro penta cycloocta deca 7, 15 dien, tetra bromo bisphenol A, tribromo phenol, tetra bromo anhydrous phthalic acid, dibromo neo pentyl glycol and the like.

As zinc compound, for instance,  $3\text{ZnO} \cdot 2\text{B}_2\text{O}_3 \cdot 3,5\text{H}_2\text{O}$ ,  $3\text{ZnO} \cdot 3\text{B}_2\text{O}_3 \cdot 3,5\text{H}_2\text{O}$ , and the like that are sulfur, zinc compound,  $\text{ZnC} \cdot \text{ZnMoO}_4$ ,  $\text{CaO} \cdot \text{ZnMoO}_4$  and the like that are moribdenum zinc compound and the like, the compound baked product of  $\text{Zn}_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}$ ,  $\text{ZnO}$  and

MgO; ZnO ZnCO<sub>2</sub> and the like can be listed.

As antimonies compounds, there are, for instance, antimony trioxide and the like.

And, considering the flexibility of the insulation material of the present invention and the purpose such as adjustability of insulation performance of the insulation body obtained from it, as the additive agents other than aforementioned, there is non polymerizable fluorine compound, for instance, fluoro alcohol like the following:

(r is an integer of 1~4, s is an integer of 1~20).

Fluoro carboxylic acid like the following:

(t is an integer or zero (0) or 1~4, u is an integer of 1~20), or fluoro polyether usually called fluorine oil; or N(C<sub>4</sub>F<sub>9</sub>)<sub>3</sub>, perfluoro decalin, C<sub>20</sub>F<sub>17</sub>O C<sub>4</sub>F<sub>9</sub>G<sub>4</sub>F<sub>20</sub> that are normally called fluorine inactive solution can be used.

And vis-à-vis the insulation material of the present invention, in order to control its viscosity, or coating characteristics, depending on needs, solvent can be added. As solvent, as long as it does not adversely impact on the polymerization reactivity, there is no limitation, for instance, methanol, ethanol, and isopropyl alcohol and that like that are alcohol type; acetone, methyl ethyl ketone, methyl isobutyl ketone and the like that are ketone type, methyl acetate, ethyl acetate, butyl acetate and the like that are ester type, chloroform, cyclo ethane, carbon tetra chloride and the

like that are chlorine type, and benzo trifluoride, chlor benzo trifluoride , m-xylene hexa fluoride, tetra chloro difluoro ethane, 1,1,2-trichlor -1,2,2,- trifluoro ethane, trichloro monofluoro methane and the like that are low boiling point solvent are preferred from the workability standpoint. And, in case solvent is contained like above, before starting the polymerization curing, normal temperature or depending on needs, using heating or depressurization, the process to remove the solvent is required. In case the solvent is heated and removed, in order for the heating polymerization of monomer and the like not to occur, it is necessary to control the temperature.

As the viscosity adjustment other than above or controlling additive agent for insulation performance , there are, for instance, acrylic type resin, fluorine type resin, polyethylene and the like that are olefin type resin, polyester type resin, polyether type resin, polysulfonic type resin, polysulfon type resin, silicone type resin, polyamide type resin, polyimide type resin, alkyd type resin, melamine type resin, urethane type resin, polyimine type resin, epoxy type resin, phenol type resin, urea type resin and the like are listed.

As coloring agent, there are, for instance, organic pigment, inorganic pigment, and organic dye and the like.

Furthermore, from the standpoint of insulation performance improvement and economy, powdery or particle shaped materials of

the following can be added: for instance, carbon silica anhydrous silica, quartz, glasses, rocks, ceramics, zircon carbonic acid calsium, magnesium oxide, calsium oxide, aluminum oxide, aluminum hydroxide, titanium oxide, antimony oxide, calsium silicate, monovalent silicon, boron nitride, moribdenum disulfide, furthermore, mica, clay, kaolin, talc, asbestos and the like.

As the method to coat the insulation material of the present invention on base material, various known methods in the industry can be used such as, for instance, brush coating, applicators, bar coater, roller brushes, or roll coaters and the like that are coating methods, the spray coating method by air spray or airless spray coating device, flow coating method by shower coater, or curtain flow coater and the like (flow coating), soaking method, casting method, and spinner coating and the like, depending on the material of the base material or shape, usage and the like, it is desired that usage is discriminated.

Particularly, in case the insulation material of the present invention is coated on electrical wire conductive bodies and insulated electrical wire is produced, preferably, the following methods are preferred; one method is that electric wire conductive body is continuously soaked in a tank that contains said insulation material, and pulled up, depending on needs, solvent is removed, then, active energy beams are irradiated, thus resin coated layer is formed; another method is that the mouth piece is designed such

that insulation material of the present invention can be fed to the mouth piece continuously, the electric wire conductive body is passed through this mouth piece and continuously coated, depending on needs, solvent is removed, then, active energy beams are irradiated, thus, insulated resin coated layer is formed.

Regarding the conductive body which can be coated by insulation material of the present invention, the entire traditional electric wire conductive bodies that are specifically limited can be used, for instance, gold wire, copper wire, copper wire plated by nickel, silver, tin and lead and the like, silver wire, nickel wires, alloy wires for electrical heating, alloy wires for electrical resistance, and iron wires and enamel wires and the like can be used. And as to its shapes, there are, for instance, round wire, straight angle wire, wires with different shapes, stranded wires and the like.

The coating film thickness to the conductive body of insulation material of the present invention differs depending on the extent of insulation characteristics needed and economic aspect, it is preferred that the cured product of said insulation material is normally 50  $\mu\text{m}$  or less, preferably 15~50 $\mu\text{m}$  in coating film thickness. The electrical wires coated by the cured product of said insulation material which is 50 $\mu\text{m}$  or less is particularly suitable for computer wire which requires fine wiring.

In case the insulation material of the present invention is polymerized and cured by active energy beams, industry wide well

known germicidal lamps, UV florescent lamps, carbon arc, xenon lamps, high pressure mercury lamps for duplication, middle pressure or high pressure mercury lamps, ultra high pressure mercury lamps, non electrode lamps, metal halide lamps, UV beams which uses natural light as light source, or electron beams by scanning type, curtain type electron beam accelerating path can be used, and in case the UV beam curing with the coating layer of 5  $\mu\text{m}$  or less in thickness is used, due to the efficiency of polymerization, it is preferred that it is irradiated in the inactive gas atmosphere such as nitrogen gas and the like.

The insulation material of the present invention requires that fluorine atoms be contained although it can be used for electronic devices, electric devices without any limitations, hence, the characteristics is that by changing its amount, the refraction rate of insulation body itself can be adjusted.

They can be used favorably as the adhesive agent and sealing agent and potting agent for optical sensors, optical IC, optical LSI that requires optimum refraction rate in addition, the optical parts which requires insulation performance. And they can be used favorably used As the sealing agent, potting agent, and adhesive agent for the electronic parts what require temperature and humidity resistance such as electronic circuit substrate, LSI, IC, relays and the like.

(Embodied example)

Next, the specific embodied examples of the present invention

will be explained; however, the present invention is not limited by such explanation of course. The [part] in the sentences are weight standard.

Hereafter, all the abbreviations of the compounds show the aforementioned compounds. The abbreviation "A" shows acrylate compound, "M" methacrylate compound, "CL" alpha- chloro acrylate compound, "CN" alpha- cyano acrylate compound.

#### Synthetic example 1 (synthesis of copolymers)

500 milliliter four mouthed flat bottom flask is equipped with a cooling condenser, a temperature gauge, a churning device, and into this flask is put in I-a-I 180g, II-2 10g, n-butyl acrylate 10g, and azo bisisobutylo nitrile (hereafter called AIBN) 1g and in N<sub>2</sub> gas atmosphere, it is churned for 30 minutes at 70 deg C. during heating, viscous mass polymerized product was taken out from the system.

The molecular weight found from GPC was Mn-450,000 by styrene computation. Fluorine atom content amount was 56.1%.

#### Synthesis example 2 (same as above).

Other than that it was made to be I-a-47/II-13(m=8)/isobornyl acrylate -60/20/20 (weight ratio), it was done same as in embodied example 1 and synthesized, consequently the styrene computed molecular weight found from GPC of this copolymer was Mn-320,000.

#### Synthesis example 3 (same as above)

500 milliliter four mouthed flat bottom flask is equipped with

a cooling condenser, a temperature gauge, a churning device, and into this flask is put in I-a-I 60g, adamantyl acrylate 5g, n-butyl acrylate 5g, thio glycolic acid octyl 0.02g, A|BN 0.3g and 1,1,1-trichloro ethane 230g and in N<sub>2</sub> gas atmosphere, it is churned for 10 hours at 80 deg C. Solvent was distilled under decompression and the targeted polymerized product was obtained.

The molecular weight found from GPC was Mn-320,000 by styrene computation.

Synthesis example 4 (same as above)

500 milliliter glass cylindere shape flask is equipped with a churner. Into this flask is put in I-a-I 372g, cyclo hexyl acrylate 28g, and starting agent V-9 0.7g and at 60 deg C, while being churned, one unit of high pressure mercury lamp 60W/cm lamp is irradiated from the side, it is reacted for 10 seconds. By this operation, viscous reaction product with viscosity 8,600cpc/25 deg C was obtained. In the reaction product, molecular weight by GPC of polymer was 110,000, and residual unreacted monomer ratio was 76% in the entire weight. And the composition ratio of copolymer of the polymer matched with the prepared monomer ratio, which was confirmed by NMR.

Synthesis example 5, 6 (same as above)

Doing same as in synthesis 4, the viscosity, of the reaction product thus synthesized, molecular weight of polymer, and ratio of the residual unreacted monomer are summarized in table 1.

Table 1

Synthesis example	Polymerization composition (%)	Viscosity (cps/25 deg c)	Residual unreacted monomer amount (%)	Molecular weight MN styrene computation
5	I-a-52/ isobornyl methacrylate 20/ 70	9.400	70	18
6	I-a-35/ I-a-33/1-a-31/t-butyl acrylate 20/40/32/8	8.100	69	41

Synthesis example 7 (same as above)

Two liter four mouthed flat bottom flask is equipped with a cooling condenser, and a churning device. Into this flask is put in methyl isobutyl ketone 120g, methaxylene hexa fluoride 120g, and its temperature is raised to 110 deg C, and 1-a-1 340g, glycidyl methacrylate 60g and L-butyl per oxy -2- ethyl hexanoate 12g are mixed, this mixture is dripped for 3 hours by using dripping funnel, furthermore reacted for one hour. Next, the homogenized mixture of t-butyl peroxy -2- ethyl hexanoate 2g and methyl isobutyl ketone 26g is dripped for one hour, furthermore, it was reacted for 7 more hours, and copolymer of molecular weight Mn-15,000 is obtained. Into this is added hydroquinone 0.2g, triethyl amine 2g and acrylic acid 32 g, was reacted for 5 hours at 110 deg c, thus fluorine containing resin was obtained that contains acryloil group on side chain with Mn=16,000, non-saturation degree 1.0. The solvent of this resin solution was distilled under decompression at 40 deg c, thus obtaining fluorine containing resin of solid portion 100%.

<Embodied example 1~38)

[Evaluation of the electrical characteristics]

Dielectric breakdown voltage, volume specific resistance rate, and dielectric rate were measured according to the method of ASTM 0149, 02457 respectively. Regarding supplied samples, insulation material described in each embodied example was irradiated and cured by one 120 W/cm high pressure mercury lamp of light converging type, and using infrared spectrometer, after confirming extinction of carbon-carbon double bonding group, it was made.

The electrical characteristics of insulation material of the present invention are shown I table 2

And, when the insulation material of the present invention was cured by heat, same electrical characteristics as the one cured by active energy beams were shown. However, compared with thermal curing, curing by active energy beams was superior in all aspect of workability, productivity and economy.

<comparison example 1~12>

Traditionally used polytetra fluoro ethylene in comparison example 7 and samples supplied in comparison example 1~6 and 8~12 were used. Other than that, comparison examples using same method as in above described embodied example were summarized and shown in table 2.

Table 2

\*1: dielectric breakdown voltage (kV.mm), \*2: volume specific resistance rate ( $\Omega$ .cm)

\*3: dielectric rate

Embodied example	Insulation material		Electrical properties		
		(parts)	*1	*2	*3
1	I-a-36	85.5	31	$2.0 \times 10^{17}$	2.0
	IV-17 (A)	11.4			
	Gamma-mercapto propyl trimethoxy silane	2.0			
	V-9	1.0			
2	I-a-35	84.0	30	$1.8 \times 10^{17}$	2.0
	IV-17 (A)	13.0			
	V-9	1.0			
3	I-a-35	15.0	27	$7.5 \times 10^{16}$	2.4
	IV-17 (A)	84.0			
	V-9	1.0			
4	I-a-35	4.3	23	$9.5 \times 10^{15}$	2.5
	IV-17 (A)	94.7			
	V-9	1.0			
5	I-a-35	56.0	40	$4.4 \times 10^{17}$	2.3
	I-b-2	30			
	V-9	2.0			
6	I-a-I	4.9	43	$6.5 \times 10^{17}$	2.1
	I-b-I	90.1			
	V-9	5.0			
7	I-a-I	4.9	23	$5.5 \times 10^{15}$	2.3
	IV-17 (A)	90.1			
	V-9	5.0			
8	I-a-I	50.0	32	$6.9 \times 10^{16}$	2.2
	IV-17 (A)	47.0			
	V-9	3.0			
9	I-a-I	93.0	34	$1.2 \times 10^{17}$	2.0
	IV-17 (A)	5.0			
	V-9	2.0			
10	I-a-I	70.0	35	$9.7 \times 10^{16}$	2.0
	Isobornyl acrylate	15.0			

	IV-17 (A)	10.0			
	V-9	5.0			
Continued page 29					
11	I-a-I	70	29	2.7 x10 <sup>14</sup>	2.0
	Z-ethyl hexyl acrylate	25.0			
	V-9	5.0			
12	I-a-I	78.9	33	1.2x 10 <sup>17</sup>	2.0
	II-13 (m=8)	18.0			
	IV-17	10.0			
	V-9	2.0			
13	I-a-I	70.0	34	1.7 x10 <sup>17</sup>	2.1
	See chemical structure page 29	19.0			
	IV-17	10.0			
	V-9	2.0			
14	I-a-I	80.0	28	3.3x 10 <sup>16</sup>	1.9
	I-a-22	30.0			
	IV-17 (A)	15.0			
	V-9				
15	CH2=CHCOOCH2CF3?	30.0	30	8.4 x 10 <sup>16</sup>	2.1
	I-a-5	50.0			
	IV-20 (A)	8.0			
	V-8				
16	I-a-I	70.0	38	2.0x 10 <sup>17</sup>	2.3
	Isobornyl acrylate	7.0			
	IV-17 (A)	8.0			
	Average particle diameter 1µm mica powder	10.0			
	V-9	5.0			
17	I-a-I	56.0	28	7.6x 10 <sup>14</sup>	2.1
	I-a-47	33.0			
	Iv-9 (A)	10.0			
	v-9	2.0			
18	VI-9	80.0	33	5.5x 10 <sup>16</sup>	2.2

	1,6- hexan diole diglycidil ether	17.0			
	X-2	3.0			
	Continued page 30				
19	VI-II	90.0	36	$7.0 \times 10^{14}$	2.1
	1,6-hexan diol diglycidil ether	7.0			
	X-2	3.0			
20	VI-II	40.0	39	$3.9 \times 10^{14}$	2.4
	VI-8	57.0			
	X-21	3.0			
21	VI-1	90.0	38	$8.7 \times 10^{14}$	2.4
	VI-2	6.0			
	X-2	4.0			
23	VI-1	70.0	31	$1.0 \times 10^{16}$	2.2
	VI-13	26.0			
	X-2	4.0			
24	VI-4	45.0	34	$3.8 \times 10^{16}$	2.1
	VI-6	50.0			
	X-2	5.0			
25	VI-II	64.0	38	$7.0 \times 10^{16}$	2.1
26	1,5-hexane diol diglycidyl ether	7.9			
	Average particle diameter 20 $\mu$ m mica	6.0			
	X-2	3.0			
	Compound in synthesis example 1	10.0	36	$7.3 \times 10^{16}$	2.2
	I-a-I	45.0			
	IV-17 (A)	42.0			
	V-5	3.0			
27	Compound in synthesis example 1	60.0	47	$7.7 \times 10^{17}$	2.6
	I-a-28	16.0			
	IV-9 (A)	20.0			
	V-9	4.0			
28	Compound in synthesis example 1	20/0	36	$2.6 \times 10^{17}$	2.0
	I-a-I	36			
	See chemical structure on page 30	14.0			
	IV-17 (A)	8.0			
	V-9	2.0			

Continued page 31					
29	Compound in synthetic example 3	10	36	1.0x 10 <sup>17</sup>	2.0
	CH2=CHCOOCH2CF3	27			
	I-a-5	54			
	IV-20 (A)	7			
	V-8				
30	Reaction production in synthetic example 4	80	37	1.5x 10 <sup>17</sup>	2.1
	IV-17(A)	15			
	V-9	5			
31	Reaction product in synthetic example 5	70	29	1.0 x 10 <sup>17</sup>	2.0
	Phenoxy ethyl acrylate	12			
	IV-8 (A) (average molecular weight 200)	12			
	V-9	6			
32	Reaction product in synthetic example 6	82	29	1.3x 10 <sup>17</sup>	2.1
	IV-17(A)	15			
	V-9				
Continued					
33	Reactive product in synthetic example 7	50	22	5.0x 10 <sup>14</sup>	2.1
	I-a-I	40			
	I-a-26	7			
	V-9	3			
34	Reactive product in synthetic example 7	30	31	9.0x 10 <sup>14</sup>	2.2
	I-a-I	39			
	I-a-47	23			
	IV-9 (A)	6			
	V-9	2			
35	Puryden polyfluoride (Molecular weight 10,000)	10	32	2.27x 10 <sup>17</sup>	2.0

	I-a-I	45			
	I-a-22	27			
	IV-17(A)	14			
	V-9				
Continued					
36	See chemical structure on page 32	86.0	33	$1.5 \times 10^{17}$	2.2
	I-a-I	10.0			
	v-9	4.0			
37	See chemical formula and chemical structure	85.0	35	$7.8 \times 10^{16}$	2.1
	I-a-15	10.0			
	V-9	4.0			
38	See chemical structure	86.0	20	$2.0 \times 10^{16}$	2.3
	VI-I	10.0			
	x-2				
Continued					
Note: wi/ means corresponding one with.					
1 wi/2	n-C <sub>11</sub> H <sub>21</sub> OCOCH=CH <sub>2</sub>	84.0	12	$7.3 \times 10^{12}$	3.3
	IV-17(A)	15.0			
	V-9	1.0			
2 wi/1	n-C <sub>11</sub> H <sub>21</sub> OCOCH <sub>2</sub> =CH <sub>2</sub>	86.5	13	$5.6 \times 10^{12}$	3.8
	IV-17(A)	11.4			
	Gamma-mercapto propyl trimethoxy silane	2.0			
	V-3	1.0			
3 wi/8	n-decyl acrylate	50.0	15	$1.1 \times 10^{13}$	3.6
	IV-17 (A)	47.0			
	V-9	3.0			
4 wi/10	n-decil acrylate	70.0	14	$7.0 \times 10^{12}$	3.7
	Isobornyl acrylate	15.0			
	IV-17 (A)	10.0			
	V-9	5			
	Continued page 33				
5 wi/13	n-decyl acrylate	70.0	13	$6.3 \times 10^{12}$	3.7
	See chemical structure on page 33	18.0			

	IV-17	10.0			
	V-9	2.0			
6 wi/18	n-decyl alcohol glycidyl ether	80.0	13.0	$1.3 \times 10^{12}$	35
	1,6-hexan diol glycidyl ether	17.0			
	x-2	3.0			
7	Polytetra fluoro ethylene		20	$1.0 \times 10^{17}$	2.1
8	I-b-2	98.0	20	$5.0 \times 10^{14}$	2.9
	V-9	2.0			
9	I-b-I	95.5	21	$7.8 \times 10^{14}$	3.0
	V-9	5.0			
	Continued				
10 wi/ 36	See chemical structure on page 33	95.0	18	$3.3 \times 10^{14}$	3.2
	v-9	4.0			
11 wi/37	See chemical structure on page 33	96.0	16	$1.2 \times 10^{14}$	3.3
	V-9	4.0			
12 wi/38	See chemical structure on page 33	96.0	19	$6.2 \times 10^{14}$	3.5
	x-2	4.0			

<Embodied example 39~75, comparison example 13-21>

(Production method of insulation coated electrical wire)

The insulation material shown in aforementioned embodied example 1~39 is fed to coating die through which external diameter 0.900 nickel plated copper round wire is placed, and after coating on said nickel plated copper round wire, UV curing furnace equipped with two output units of 120W/cm metal halide lamps is filtered, thus obtaining coated electrical wire with external diameter of 0.980mm.

Regarding the coated electrical wire of the thermally melting type fluorine resin as shown in figure 7, fluorine type resin is introduced into the extruder with the following temperature and

diameter, it was extruded so that aforementioned conductive body with external diameter 0.900 mm was coated to have external diameter 0.980 mm.

[barrel band (zone) 1 temperature is 260 deg C, and barrel zone 2 temperature is 270 deg c, barrel zone 3 temperature is 300 deg c, head temperature is 320 deg c, die part temperature is 320 deg c, 49mm L/D=20.]

As clarified by above described condition, thermally melting type fluorine resin has high melting temperature, and compared with the active energy heat curing of the insulation material of the present invention; it is inferior in all aspects of workability, productivity and economy.

And, in table 3, the number of insulation material shows the resin composition which corresponding with the number of embodied example in table 2. And regarding the line speed, it shows the production speed (wire drawing coating speed) per one resin coating wire. Furthermore, the external appearance of resin coated electrical wire follows JIS c2358, and evaluation of pinholes followed JIS c3203.

Table 3

Embodied example	Insulation material	Line speed (m/minute)	Outer appearance	Pin hole
39	Embodied example : product 1	20	Good	None
40	Same: same 1	150	Same	Same
41	Same: same 1	250	Same	Same

42	Same: same 2	200	Same	Same
43	Same: same 3	200	Same	SAME
44	Same: same 4	200	Same	Same
45	Same: same 5	200	Same	Same
46	Same: same 6	200	Same	Same
47	Same: same 7	200	Same	Same
48	Same: same 8	200	Same	Same
49	Same: same 9	200	Same	Same
50	Same: same 10	200	Same	SAME
51	Same: same 11	200	Same	SAME
52	Same: same 12	200	Same	Same
53	Same: same 13	200	Same	Same
54	Same: same 14	200	Same	Same
55	Same: same 15	200	Same	Same
Continued				
56	Same: same 17	250	Same	Same
57	Same: same 18	150	Same	Same
58	Same: same 19	150	Same	Same
59	Same: same 20	100	Same	Same
60	Same: same 23	150	Same	Same
61	Same: same 24	150	Same	Same
62	Same: same 25	150	Same	Same
63	Same: same 26	300	Same	Same
64	Same: same 27	Same	Same	Same
65	Same: same 28	Same	Same	Same
66	Same: same 29	Same	Same	Same
67	Same: same 30	Same	Same	Same
68	Same: same 31	Same	Same	Same
69	Same: same 32	Same	Same	Same
70	Same: same 33	Same	Same	Same
71	Same: same 34	Same	Same	Same
72	Same: same 35	Same	Same	Same
Continued				
73	Same: same 36	100	Same	Same
74	Same: same 37	Same	Same	Same

75	Same: same 38	Same	Same	Same
Comparison example				
13	Comparison example product 2	150	Defective	Many
14	Same: same 6	80	Same	Same
15	Polytetra fluoro ethylene	5	Same	Same
16	Tetra fluoro ethylene. propylene copolymer	5	Good	None
17	Same	10	Defective	Many
18	Comparison example product 10	100	Same	Same
19	Same: same 10	80	Same	Same
20	Same: same 11	100	Same	Same
21	Same: same 12	100	Same	same

<Embodied example 76>

Regarding the insulation material of embodied example 31 (after curing, the light permeation rate of 1mm thick chip was 92% (850nm), refraction rate  $n=1.465$ ) is potted into the optical IC chip which is supported on the glass substrate, and is irradiated by one 80 W/cm high pressure mercury lamp for two seconds, and cured. Potted optical IC chip was maintained in a vessel a constant temperature and constant humidity at 70 deg C 95 % for 1,000 hours, there was no floating peeling in the potted part, no corrosion was observed in the circuit. Furthermore, as the performance of optical IC chips, it was by far superior to that of the optical IC chip sealed by traditional potting agent.

[Effects of the invention]

The curable insulation material of the present invention contains polyfluoroalkyl group - containing mono(meta) acrylate monomer or polyfluoroalkyl group - containing monoepoxide monomer; hence, compared with the cured product of traditional curable insulation material, the insulation performance is greatly superior.

Hence, the insulation material of the present invention is favorable as the insulation coated electric wires, insulation sealing material, electrical insulation layered plate material.